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NOVEL CHEMICAL LASER SYSTEMS.(U)  
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20. ABSTRACT (Continued)

from 1.152 to 14.54 microns and were identified with transitions in the atoms: As, Bi, Cd, Fe, Ga, Ge, Hg, In, Ni, Pb, Sb, Se, Sn, Te, Tl, V and Zn. Electronically excited YCl was formed from the reaction of chlorine with Y metal; chemiluminescent spectra from the excited YCl species were recorded in a shock tube-driven supersonic mixing flame. Strong emission was noted near 4000Å and weaker bands at longer wavelengths believed to arise from triplet states of chemically formed YCl. This reaction system is of interest because of a high photon yield, a high degree of specificity in energy disposal among product states and the possible existence of laser transitions of appropriate radiative lifetimes for chemical laser operation. Electrical discharges in PH<sub>3</sub>/N<sub>2</sub>O leads to strong visible chemiluminescence from an excited (P<sub>0</sub>)<sub>2</sub> species. The chemiluminescence is continuous from about 3200Å to beyond 2 micron with a broad maximum near 7500Å. The emission is believed to arise from the (P<sub>0</sub>)<sub>2</sub> excimer and may lead to a chemically pumped laser capable of broad band tuning throughout the visible spectral region.

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FINAL SCIENTIFIC REPORT

NOVEL CHEMICAL LASER SYSTEMS

Air Force Office of Scientific Research

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For The Period June 15, 1974 Through March 31, 1977

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**AIR FORCE OFFICE OF SCIENTIFIC RESEARCH (AFSC)**  
**NOTICE OF TRANSMITTAL TO DDC**

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**A. D. BLOSE**  
**Technical Information Officer**

## I. INTRODUCTION

During the past 3 years a large number of reactions have been studied at several laboratories as possible candidates for electronic transition chemical lasers. At present only two or three reaction systems appear to provide the high photon yields, the desired specificity of excitation of product states, and reaction products of suitably long radiative lifetimes. For this reason we have chosen to study the  $Y + Cl_2 \rightarrow YCl^* + Cl$  atom-exchange reaction and a second reaction system which leads to chemically formed  $(PO)_2^*$  excimer molecules. Sections II and III of this report summarize the present status of studies of the yttrium chloride ( $YCl$ ) molecular system; Section IV discusses work on a potentially efficient chemically pumped  $(PO)_2^*$  excimer system. This report is intended to summarize progress made under AFOSR Grant 73-2550 for the period June 15, 1974 through March 31, 1977. Research on these reaction systems is being continued under AFOSR Grant 77-3358.

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## II. SHOCK TUBE STUDIES OF THE $Y + Cl_2 \rightarrow YCl^* + Cl$ REACTION

Tables I and II summarize some recent photon yield measurements for several reactions which produce diatomic metal oxide or metal halide reaction products. Most of the reactions for which data are available, both at high and low pressures, exhibit a low photon yield at low pressure (ca.  $10^{-3}$  Torr) and a high yield at high pressures (ca. 1 Torr). In some cases the photon yields are remarkably high (e.g., SmF and SnO), exceeding 50% at high pressures. In general, these reactions do not produce a high degree of specificity of excitation either at low or high pressures. The chemiluminescence from  $Sm + O_3$ , illustrated in Figure 1, indicates the typically broad featureless emission observed for many of the reactions investigated to date. These reactions include Sm, Eu, and Yb with oxidizers  $Cl_2$ ,  $F_2$ ,  $N_2O$ ,  $O_3$ , and  $NO_2$ .<sup>1,2</sup> Spectra observed at high pressures are very similar to the low pressure spectra for these systems.

The most extensively studied reactions to date are the  $Ba + O_3 \rightarrow BaO + O_2$  and  $Ba + N_2O \rightarrow BaO + N_2$  reactions. These reactions give similar spectra. At high pressures the emission is dominated by  $A^1\Sigma^+ \rightarrow X^1\Sigma^+$  bands which are responsible for the high photon yields of Table II. Careful studies by several groups, Field, et al.,<sup>3</sup> Palmer, et al.,<sup>4</sup> and Eckstrom, et al.,<sup>5</sup> have established that the  $A^1\Sigma^+ \rightarrow X^1\Sigma^+$  emission results from energy transfer from one or more precursor states. The  $BaO(a^3\pi)$  state and high vibrational levels of the  $BaO(X^1\Sigma^+)$  state have been suggested as such possible primary reservoir states for the energy release of chemical reaction.

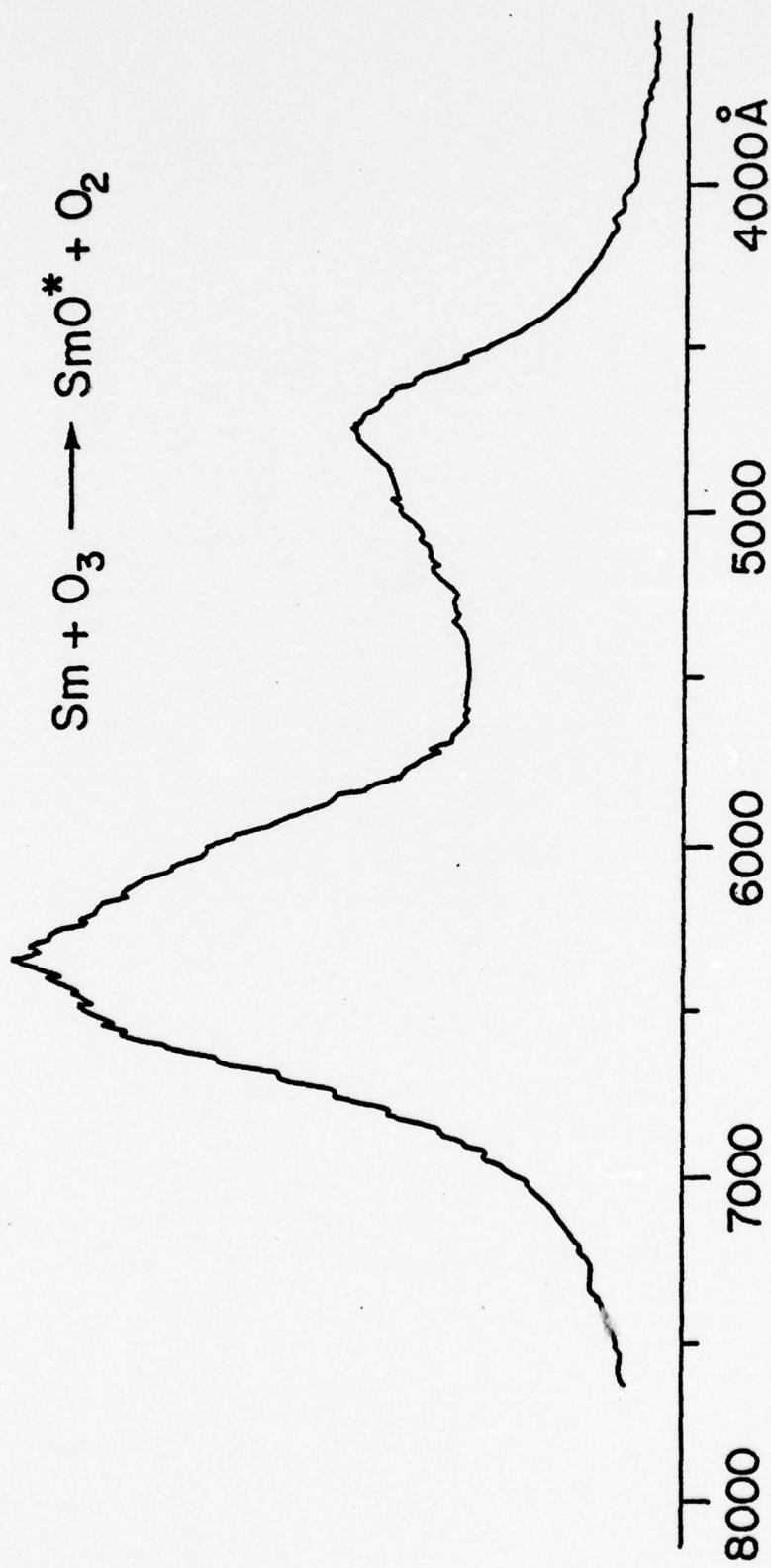


Figure 1: Visible Chemiluminescence from the Reaction  $\text{Sm} + \text{O}_3 \rightarrow \text{SmO}^* + \text{O}_2$  Under Single Collision Conditions. C.R. Dickson and R.N. Zare, Chem. Phys. 7, 361 (1975).



TABLE I  
SUMMARY OF SOME RECENT (%) PHOTON YIELD MEASUREMENTS  
UNDER SINGLE COLLISION CONDITIONS (ca.  $10^{-4}$  Torr)

<u>Metal</u>	<u>Oxidizers</u>				
	<u>N<sub>2</sub>O</u>	<u>NO<sub>2</sub></u>	<u>O<sub>3</sub></u>	<u>F<sub>2</sub></u>	<u>Cl<sub>2</sub></u>
Ba	2.3 <sup>b</sup>	0.2 <sup>b</sup>	--	--	--
Eu	0.2 <sup>a</sup>	0.007 <sup>a</sup>	0.3 <sup>a</sup>	0.9 <sup>a</sup>	--
Sc	--	--	--	$\geq 10^c$	--
Sm	0.3 <sup>a</sup>	0.07 <sup>a</sup>	0.9 <sup>a</sup>	11.8 <sup>a,b</sup>	--
Y	--	--	--	--	$\geq 5^c$

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<sup>a</sup> C.R. Dickson and R.N. Zare, Chem. Phys. 7, 361 (1975).

<sup>b</sup> C.R. Dickson, S.M. George, and R.N. Zare, J. Chem. Phys. 67, 1524 (1977).

<sup>c</sup> J.L. Gole, Electronic Transition Lasers II, L.E. Wilson, S.N. Suchard, and J.I. Steinfeld, Editors, Proceedings of the Third Summer Colloquium on Electronic Transition Lasers, Snowmass-in-Aspen, Colorado, Sept. 1976. (MIT Press, 1977) pp. 136-165.

TABLE II  
SUMMARY OF SOME RECENT (%) PHOTON YIELD MEASUREMENTS  
AT HIGH PRESSURES (ca. 1 Torr)

<u>Metals</u>	<u>Oxidizers</u>						
	<u>N<sub>2</sub>O</u>	<u>NO<sub>2</sub></u>	<u>O<sub>3</sub></u>	<u>O<sub>2</sub></u>	<u>F<sub>2</sub></u>	<u>NF<sub>3</sub></u>	<u>Cl<sub>2</sub></u>
Ba	25 <sup>a</sup>	--	36 <sup>a</sup>	8 <sup>a</sup>	4.4 <sup>a</sup>	50 <sup>a</sup>	0.6 <sup>c</sup>
Ca	--	--	--	--	0.3 <sup>c</sup>	--	--
Eu	20 <sup>a</sup>	--	5 <sup>a</sup>	2.5 <sup>a</sup>	2.5 <sup>a</sup>	--	--
Ge	0.15 <sup>b</sup>	0.05 <sup>b</sup>	--	0.2 <sup>b</sup>	0.07 <sup>b</sup>	--	10 <sup>-3b</sup>
Mg	--	--	--	--	0.02 <sup>c</sup>	--	--
Sm	38 <sup>a</sup>	--	8 <sup>a</sup>	9 <sup>a</sup>	64 <sup>a</sup>	70 <sup>a</sup>	--
Sn	6.7 <sup>d</sup> (50 <sup>e</sup> )	--	--	0.08 <sup>d</sup>	0.3 <sup>d</sup>	--	<10 <sup>-2d</sup>
Sr	--	--	--	--	0.1 <sup>c</sup>	--	--

<sup>a</sup>D.J. Eckstrom, S.A. Edelstein, D.L. Huestis, B.E. Perry, and S.W. Benson, J. Chem. Phys. 63, 3828 (1975).

<sup>b</sup>G.A. Capelle and J.M. Brom, Jr., J. Chem. Phys. 63, 5168 (1974).

<sup>c</sup>D.J. Eckstrom, S.A. Edelstein, and S.W. Benson, J. Chem. Phys. 60 2930 (1974).

<sup>d</sup>G.A. Capelle and C. Linton, J. Chem. Phys. 65, 5361 (1976).

<sup>e</sup>W. Felder and A. Fontijn, Chem. Phys. Lett. 34, 398 (1975).

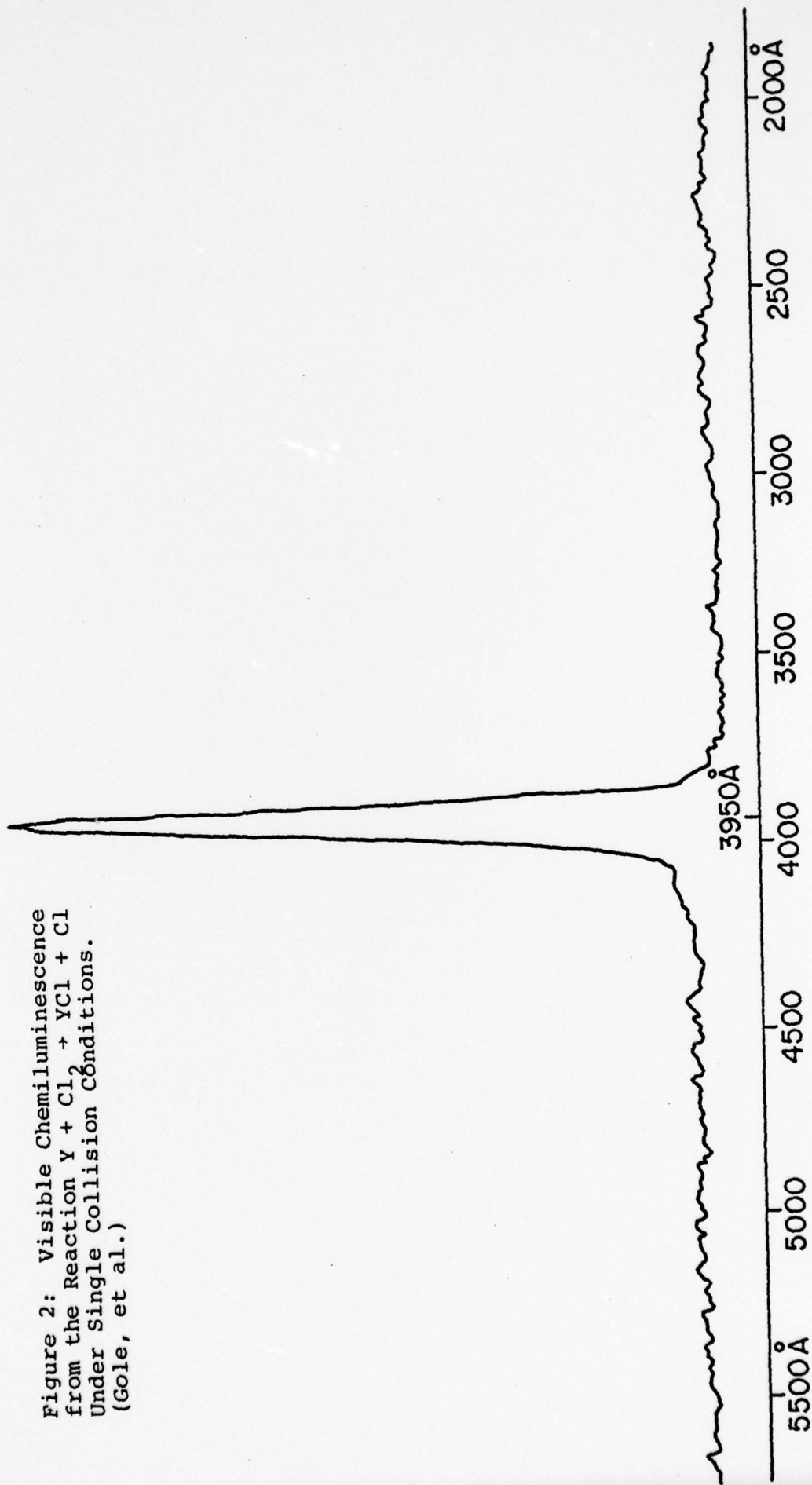
The several reactions of Tables I and II which exhibit a strongly pressure-dependent photon yield apparently conform to a simple mechanistic explanation.<sup>1</sup> A large fraction of the energy release of reaction is divided in a nearly statistical fashion among the various accessible internal product states. These states include high vibrational states of the ground electronic state and somewhat lower vibrational states of several electronically excited states. Communication between these electronic and vibrational states is maintained by rapid collisional processes. At low pressures the primary contributions to the photon yield come from directly excited states with relatively short radiative lifetimes. At higher pressures electronic states of long radiative lifetimes and high vibrational levels of the ground state are coupled to one or more electronic states of short radiative lifetimes to provide an efficient path for radiation as a major energy loss mechanism.

In contrast to all other reactions studied to date, the reactions forming ScF and YCl have a high degree of specificity and high photon yields under single collision conditions. Chemiluminescent spectra observed by Gole, et al.,<sup>6,7</sup> for the  $Y + Cl_2 \rightarrow YCl^* + Cl$  reaction are shown in Figure 2. The narrow emission feature near 3950 Å from  $YCl^*$  is thought to arise from transitions from a  $^3\Sigma^+$  upper state to the  $X^1\Sigma^+$  ground state. Although very little is presently known concerning electronic states in YCl, the low-lying electronic energy levels for ScF should be similar; these are indicated in Figure 3. Fairly complete spectroscopic analysis of the singlet bands  $E^1\pi \rightarrow X^1\Sigma^+$ ,  $C^1\Sigma^+$ , and  $B^1\pi \rightarrow X^1\Sigma^+$  exists.<sup>8</sup>

# CHEMILUMINESCENT SPECTRUM



Figure 2: Visible Chemiluminescence  
from the Reaction  $Y + Cl_2 \rightarrow YCl + Cl$   
Under Single Collision Conditions.  
(Gole, et al.)





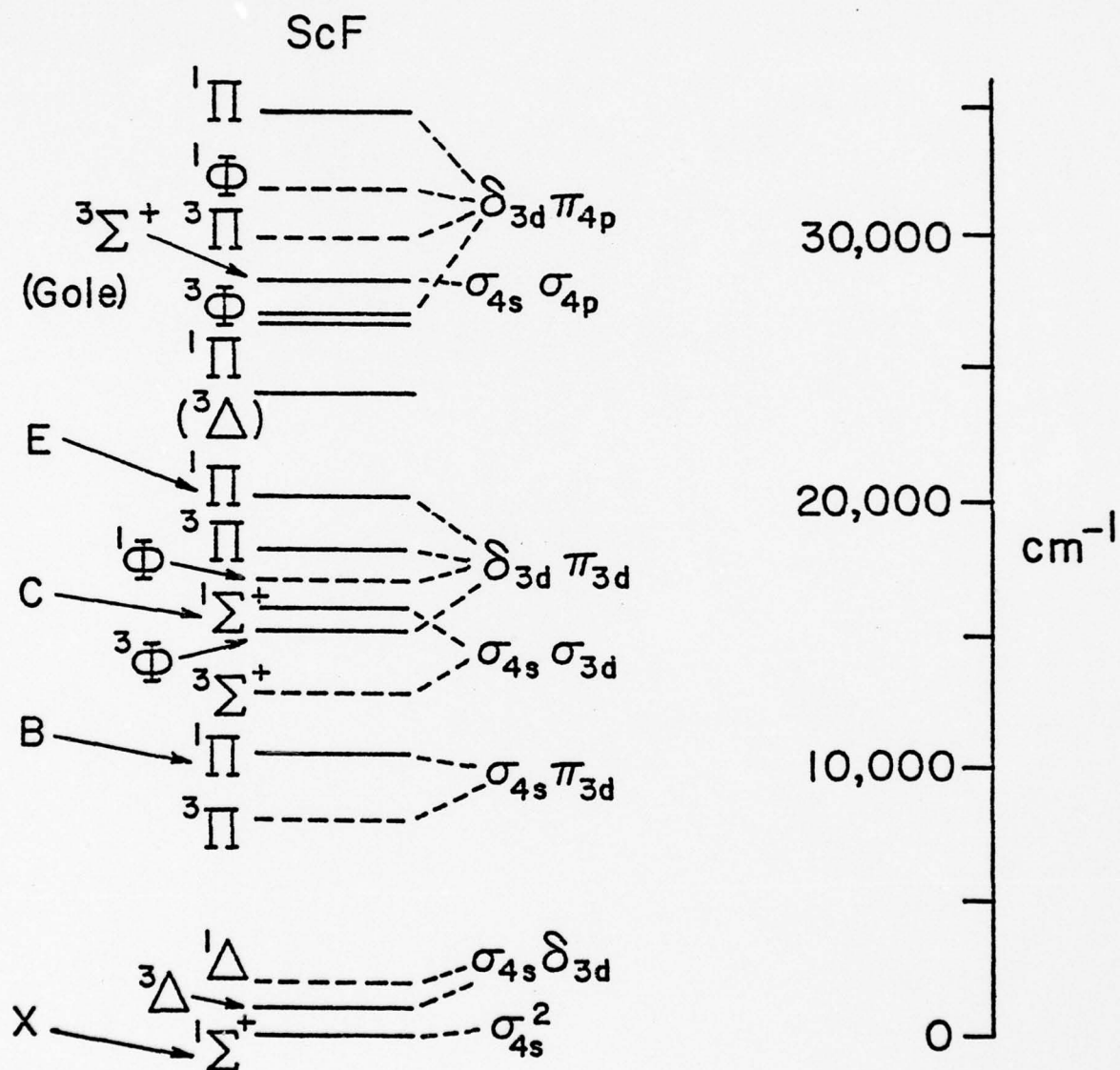


Figure 3: Low-lying Electronic Energy Levels for ScF (adapted from C.J. Cheetham and R.F. Barrow, *Advances High Temp. Chem.* 1, 7 (1967)).

The  $^3\Sigma^+ \rightarrow X^1\Sigma^+$  transition near 350 Å observed by Gole, et al.,<sup>6,7</sup> may be the band previously observed at 3360 Å in a neon matrix at 4°K.<sup>9</sup> The molecular orbitals to which the two 4s electrons in the Sc atom are promoted are given for several of the excited states of ScF in Figure 3. At present the only measured radiative lifetime for ScF bands is that of the  $E^1\pi \rightarrow X^1\Sigma^+$  band which is approximately  $1.1 \times 10^{-4}$  sec.<sup>8</sup> The lifetime for the  $C^1\Sigma^+ \rightarrow X^1\Sigma^+$  band is probably shorter, on the order of  $10^{-5}$  sec. As illustrated in Figure 3, low-lying molecular orbits of ScF are similar in character to corresponding atomic orbitals in Sc.<sup>10</sup> Transitions between electronic states in ScF may be thought of as essentially two-electron transitions between atomic orbitals in Sc. Such transitions are of  $sd \rightarrow ss$ ,  $dd \rightarrow ss$ ,  $dd \rightarrow sd$ , for  $sd \rightarrow sd$  character, and would not be expected to have large transition probabilities. It seems likely that most of the transitions in the singlet and triplet manifolds of ScF have radiative lifetimes of the order of  $10^{-3}$  to  $10^{-5}$  seconds.

These considerations suggest that the reaction of Figure 2 has the desired specificity and leads to reaction product states of long radiative lifetimes. A third desirable characteristic of these reactions, a high photon yield, has been confirmed in recent studies of Gole and co-workers.<sup>7</sup> Their measurements indicate a photon yield of 4.8%, under single collision conditions, for the 3500 Å  $^3\Sigma^+ \rightarrow X^1\Sigma^+$  band of ScF\* and a comparable photon yield for the corresponding band of YCl\* at 3950 Å. Even though, at present, nothing is

known concerning the collisional quenching of  $\text{ScF}^*$  and  $\text{YCl}^*$ , these molecules appear to be favorable candidates for electronic transition chemical lasers.

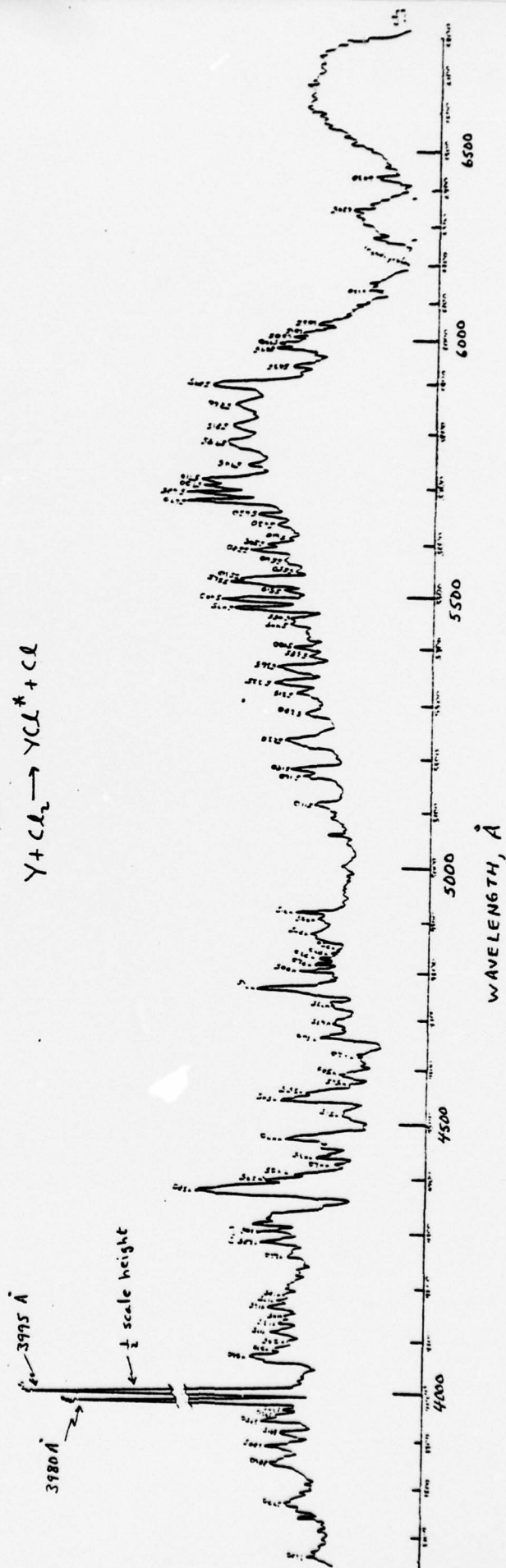
Figure 4 schematically illustrates the experimental apparatus we employ for studies of the reactions  $\text{Sc} + \text{F}_2 \rightarrow \text{ScF}^* + \text{F}$  and  $\text{Y} + \text{Cl}_2 \rightarrow \text{YCl}^* + \text{Cl}$ . A heated section of the shock tube permits vaporization of either  $\text{ScCl}_3$  or  $\text{YCl}_3$  in the presence of an argon diluent. The heated gases are initially confined between thin metal diaphragms which are ruptured by the passage of the shock wave. Shock wave heating accomplishes dissociation of the  $\text{ScCl}_3$  or  $\text{YCl}_3$  before the gases are accelerated through a supersonic nozzle array. This primary flow is then mixed with a secondary flow of either  $\text{F}_2$  or  $\text{Cl}_2$  through slots at the trailing edge of each nozzle blade. The desired reactions are initiated in the supersonic mixing zone immediately downstream of the nozzle array. The pressures and temperatures in this zone are typically about 5-20 Torr and 800-1200°K, respectively.

Figures 5 and 6 present chemiluminescence spectra for the  $\text{Y} + \text{Cl}_2 \rightarrow \text{YCl}^* + \text{Cl}$  reaction observed over the wavelength interval 3700-6700 Å. The spectra are uncorrected for film response characteristics. Predominant features of the spectra are the intense bands at 3980 and 3995 Å which are believed to correspond to the emission peak recorded by Gole et al., near 3950 Å shown in Figure 2. The data of Gole, et al, were taken under single collision conditions at pressures of about  $10^{-4}$  Torr. In contrast the data of Figures 5 and 6 were obtained at a pressure of about 10 Torr.

Figure 5: Shock Tube Chemiluminescence Data.

# CHEMILUMINESCENCE SPECTRA

For





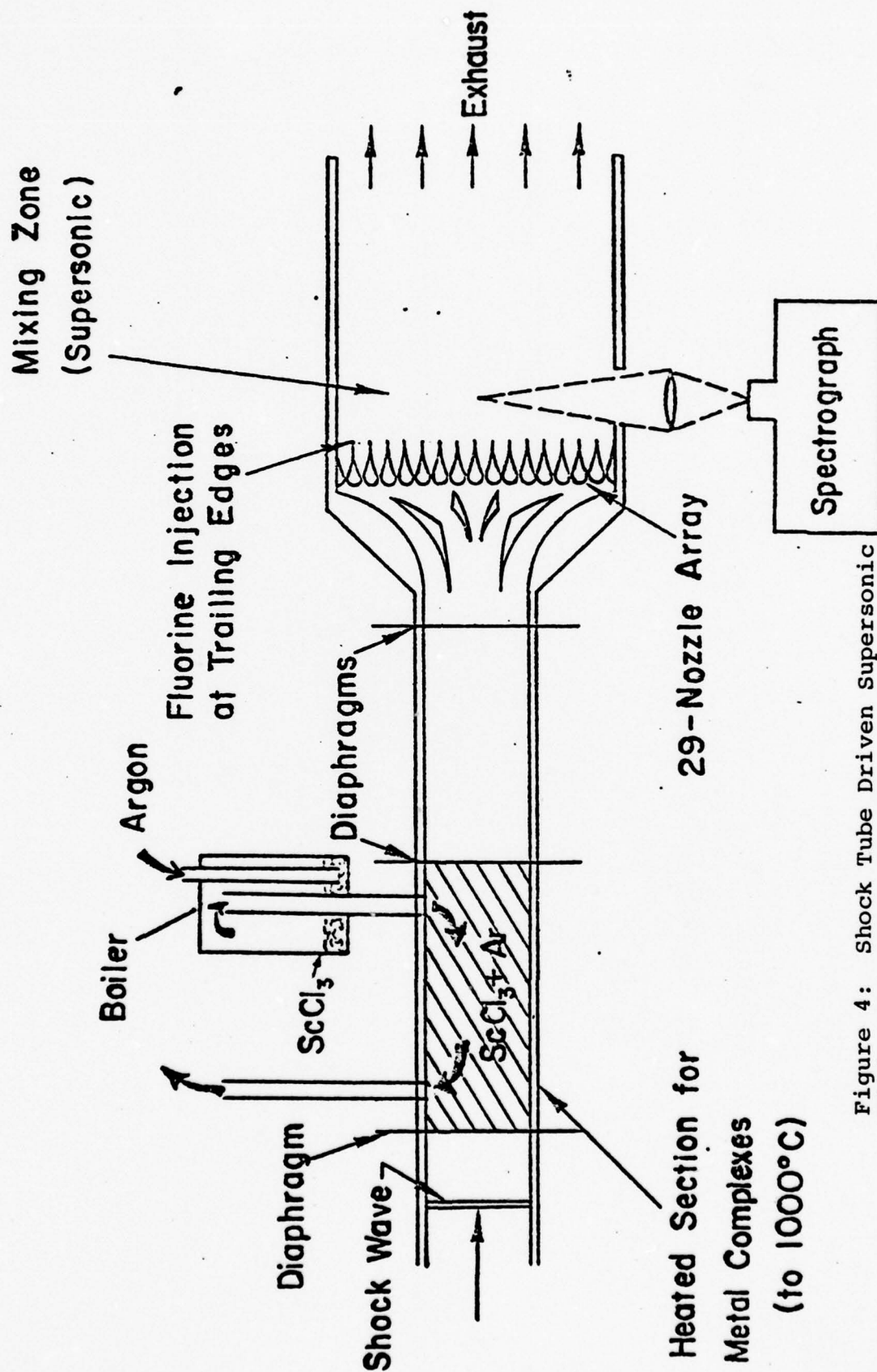
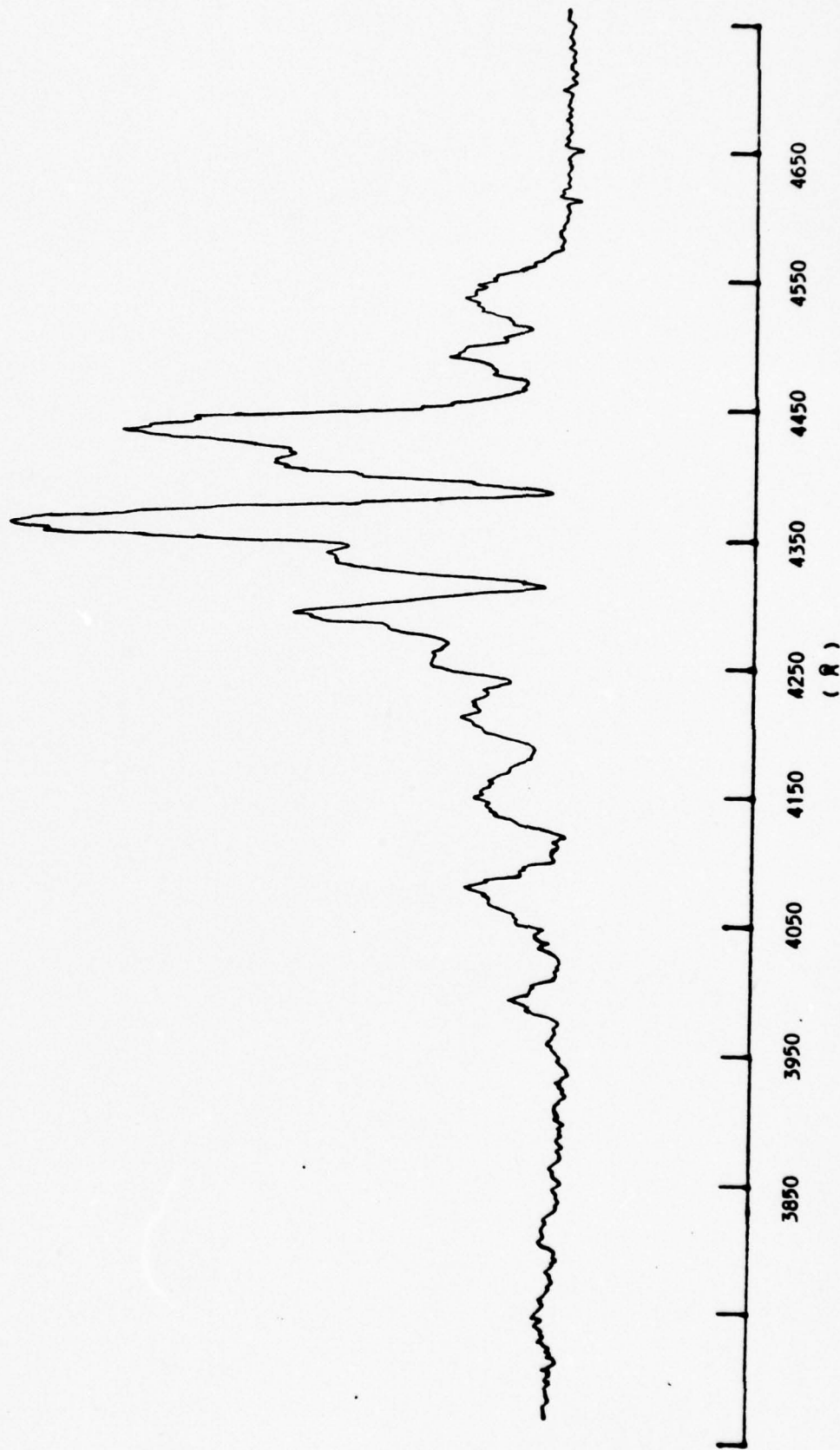


Figure 4: Shock Tube Driven Supersonic Mixing Apparatus for Study of the Reaction  $\text{Sc} + \text{F}_2 \rightarrow \text{ScF} + \text{F}$  and  $\text{Y} + \text{Cl}_2 \rightarrow \text{YCl} + \text{Cl}$ .

Figure 6: Shock Tube  
Chemiluminescence Data.

\* Spectra  
YCI  
4250 Å Band Center  
x 5 Scale



The complex band sequences at longer wavelengths have yet to be analyzed, but are believed to be both singlet-singlet and triplet-triplet bands of YCl. Figure 6 indicates a set of bands near 4300 Å which may be  $^3\Pi \rightarrow ^3\Delta$  or  $^3\Delta \rightarrow ^3\Delta$  bands. To our knowledge none of the bands of Figures 5 and 6 have been previously observed.

It seems likely that direct chemical formation of the  $^3\Sigma^+$  state as suggested by Gole, et al., is a precursor to the appearance of the longer wavelength bands we observe at high pressure. These additional bands may be excited by energy transfer processes originating from the  $^3\Sigma^+$  state. If this situation in fact exists the prospect for population inversions among several states situated below the  $^3\Sigma^+$  state appear to be good.

Additional experiments are currently underway with the shock tube apparatus of Figure 4. Attempts are in progress to observe laser action in the near infrared part of the spectrum. An optical cavity consisting of hole-coupled gold-coated mirrors oriented transversely to the flow direction is used. A Ge-Cu high speed infrared detector with spectral sensitivity from about 1-25 microns is employed to detect laser output.

Potential laser transitions include triplet-triplet, singlet-singlet, and triplet-singlet transitions at wavelengths beyond 1  $\mu$  and vibration-rotation transitions for the ground electronic state near 25 microns.

### III. LASER INDUCED FLUORESCENCE STUDIES OF YCl

Much additional information is needed to evaluate the potential of YCl as a chemical laser molecule. Spectral analysis of only one band system of YCl is currently available<sup>8</sup> and molecular constants have only been determined for the ground state and one electronically excited state. As many as 6 additional excited states may be identified from complete analysis of the spectra presented in Figures 5 and 6. The energies and identities of these states need to be determined. Moreover, radiative lifetimes and collisional quenching data are necessary for these several states.

The question of how energy is coupled by collisional processes from the nascent chemically pumped states to other radiating states must be answered before estimates of laser performance can be made.

In an effort to determine the molecular properties of excited electronic states in YCl we have designed and constructed apparatus shown schematically in Figures 7 and 8. A tungsten crucible located within a graphite furnace is heated to a temperature of 2200°K to provide an effusive flow of yttrium vapor. Molecular chlorine is injected into the primary metal atom flow by means of a water cooled injector located 3 cm downstream from the effusive orifice. Pressures within the mixing and reaction zone can be varied from  $10^{-3}$  to  $10^{-1}$  Torr.

A pulsed tunable dye laser pumped by a 200 kW nitrogen laser at 10 Hz is used for excitation of molecular bands in



# LASER INDUCED FLUORESCENCE IN A MIXING FLAME

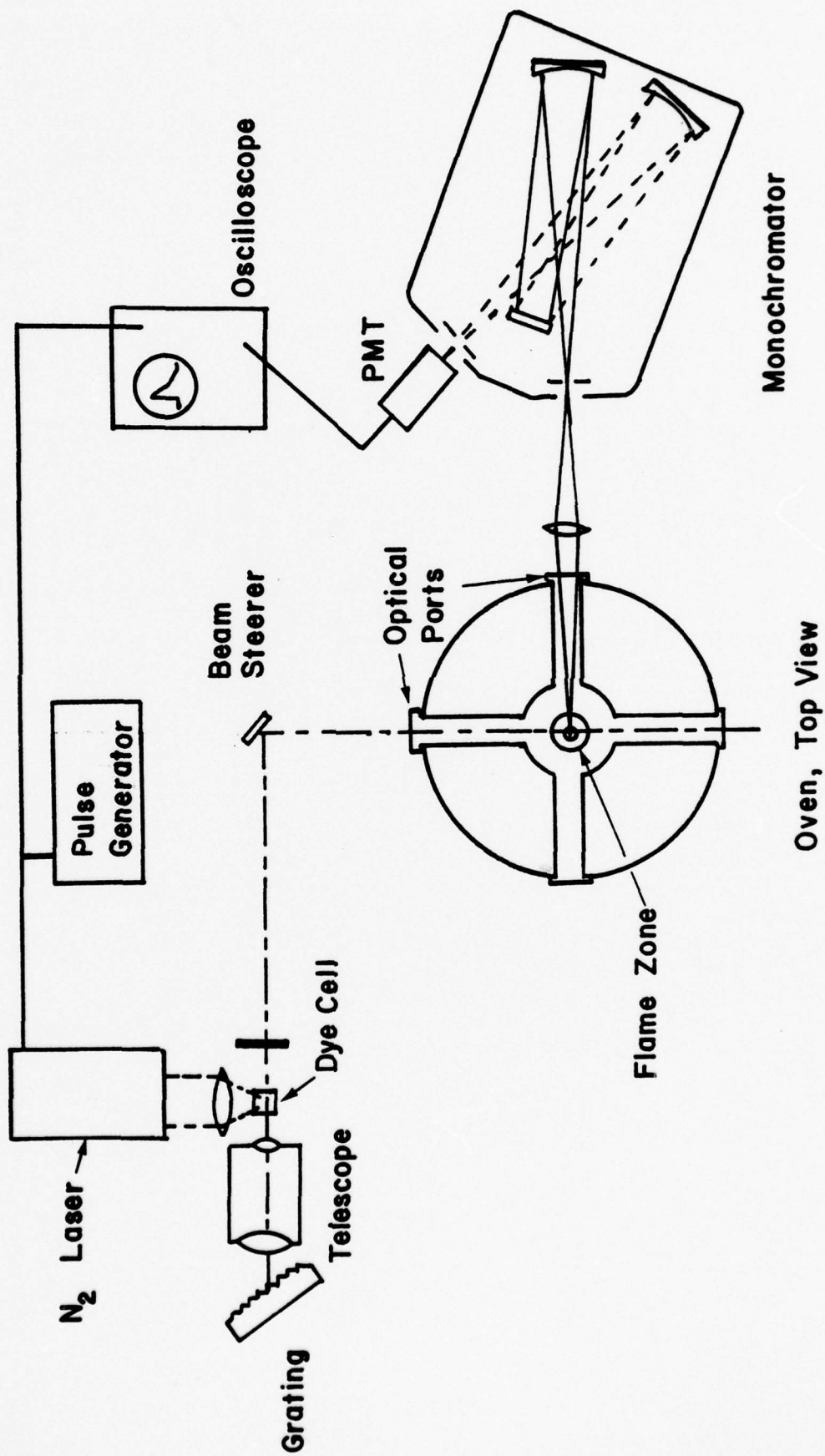


Figure 7

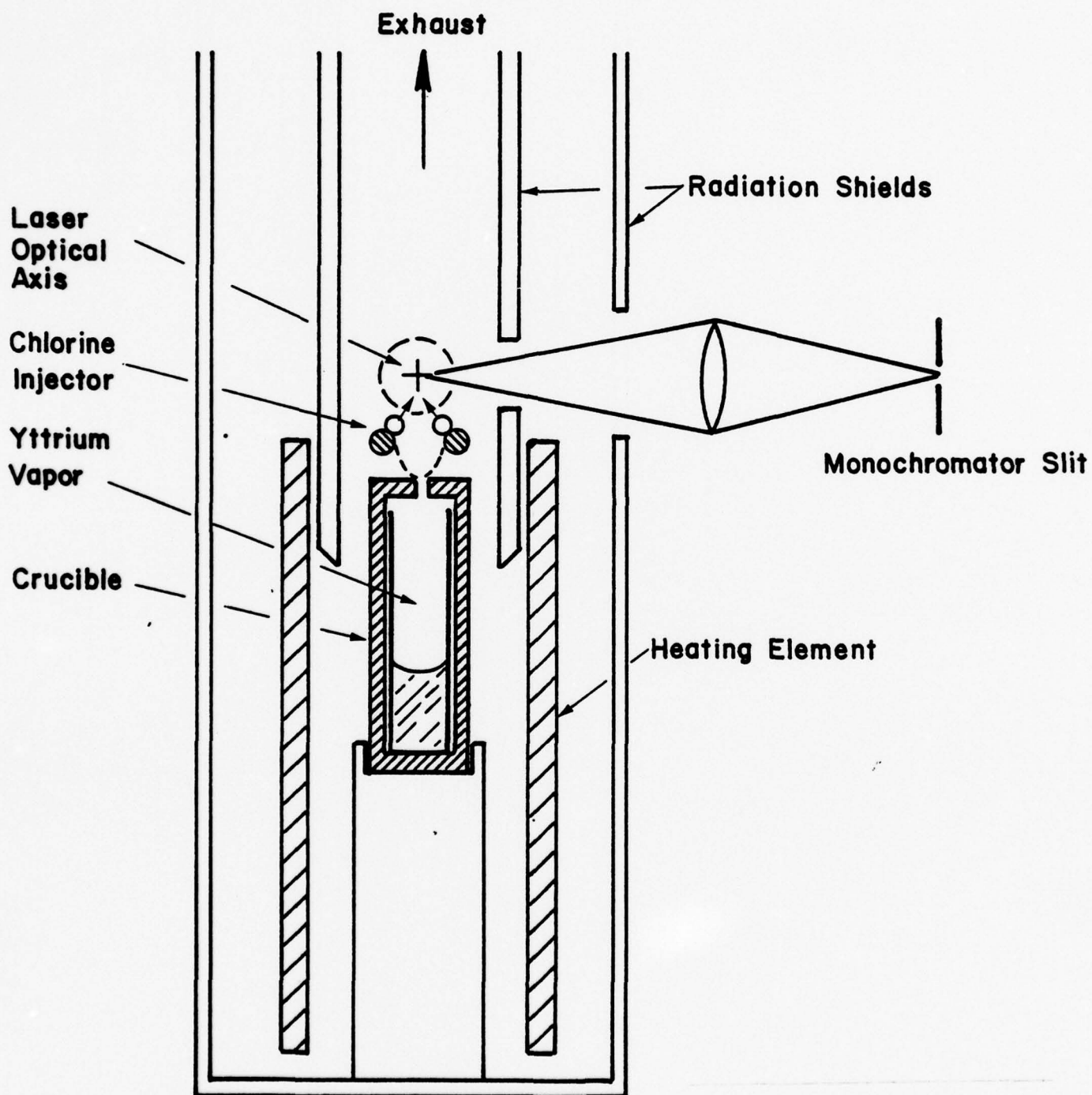


Figure 8: Flow Geometry for YCl Mixing Flame.

chemically formed YCl. Laser energies of about 10-20  $\mu$ J are available with use of various dyes covering the wavelength region from 4000-6000 Å. The dye laser is a conventional design with grating and telescope capable of 0.4 Å bandwidth in this spectral range.

The laser intensity and fluorescence signal are monitored with PM tubes. The PM tube signals are fed into a dual channel boxcar integrator.

At present the apparatus is not completely operational because of some difficulties we encountered with the oven flow system. When modifications are completed we plan to make several types of measurements. The dye laser will be progressively tuned from 3800 to 6000 Å and excitation spectra from YCl recorded. The high resolution and reproducibility afforded with this method should permit identification of many of the YCl molecular bands (Figs. 5 and 6) observed in the shock tube studies.

Radiative lifetimes will be determined for selected molecular bands with measurements of the fluorescence decay times at low reaction zone pressures. Additional fluorescence decay measurements will be made as a function of the pressure of several additive gases to determine collisional quenching cross sections for YCl states of laser interest.

In some cases it may also be possible to measure energy transfer rates between states. An example of great interest would be a determination of energy transfer rates from the upper states of the 3980 to 3995 Å bands of Figure 5 to other

lower-lying states. The tunable dye laser affords the possibility of selective excitation of the 3980 and 3995 Å bands. A search will be made for fluorescence from other bands at longer wavelengths as the pressure of the reaction zone is progressively increased. Measurements of energy transfer rates will require observations of the decay times of the 3980 and 3995 Å fluorescence and fluorescence rise and decay times for the longer wavelength secondary fluorescences.

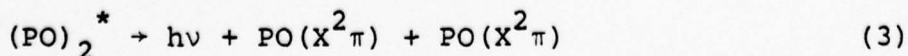
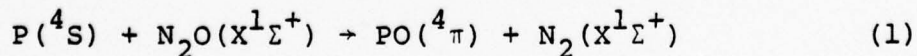


#### IV. AN IDEAL REACTION MECHANISM FOR AN EFFICIENT VISIBLE CHEMICAL LASER

The remarkably high photon yields observed for several of the reactions of Table II hold much promise for efficient chemical laser operation. These reactions demonstrate that radiant energy can be efficiently obtained from chemical reactions even when the released energy initially resides among nonradiating states. What is needed is a kinetic mechanism which acts to ensure that this radiant energy is emitted from the upper state of an inverted population.

An ideal mechanism of this type is illustrated in Figure 9. Given a chemically produced metastable atom or molecule  $A^*$ , suppose that an association reaction  $A^* + BC \rightarrow (ABC)^*$  can occur to form an electronically excited excimer molecule  $(ABC)^*$ . If radiative transitions to a repulsive energy hypersurface account for the major source of decomposition of the excimer, then efficient laser action may be possible analogous to that of the recently developed rare gas-halide ultraviolet lasers.

An example of such a system which we are currently investigating consists of the reaction steps



# A CHEMICALLY PUMPED EXCIMER LASER

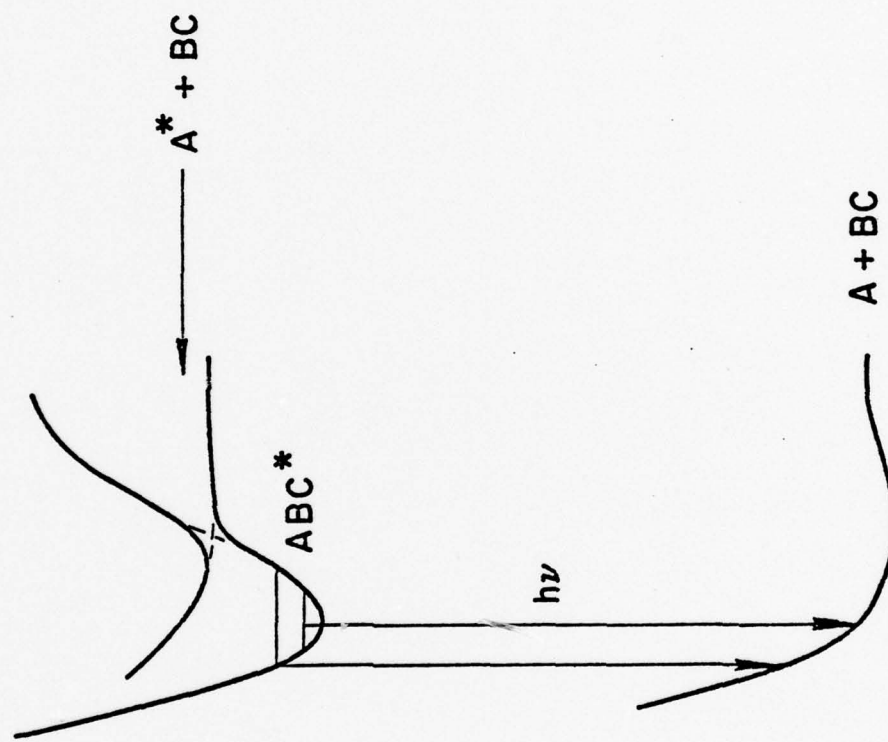


Figure 9

Reaction (1) is expected to lead to the efficient production of the metastable  $\text{PO}(^4\pi)$  state illustrated in Figure 10. Precursor reactions of importance in the  $\text{PH}_3/\text{N}_2\text{O}$  system are listed in Table III. Studies of the chemiluminescence resulting from the discharge initiation of chemical reactions in mixtures of  $\text{PH}_3$  and  $\text{N}_2\text{O}$  have revealed a strong continuous emission extending from about 3200 Å to beyond 2 μ with a broad maximum near 7500 Å. Chemiluminescence spectra are presented in Figures 11-13. The spectra of Figures 11 and 12 are uncorrected for film response. This emission is believed to arise from the  $(\text{PO})_2^*$  excimer emission of process (3), as indicated schematically in Figure 14. To date attempts to achieve laser emission from this system have been inconclusive, although photon yields for wavelengths from 3200 Å to 2 μ have been found to exceed  $10^{-3}$  under favorable conditions.

A flashlamp pumped dye laser has been used to measure the intracavity absorption spectra associated with the  $\text{PH}_3/\text{N}_2\text{O}$  emission over the wavelength range from 5000-6500 Å. Typical pulse shapes for the dye laser and excimer emission are given in Figure 15. Very little absorption occurs when the two profiles coincide as is the case in Figure 15. A strong molecular absorption is observable, however, when the dye laser is delayed with respect to the excimer emission.

Strong evidence in favor of the excimer hypothesis is given in Figure 16. Here the peak spontaneous emission intensity determined from emission profiles similar to the





TABLE III  
Discharge-Initiated Reactions  
in  $\text{PH}_3/\text{N}_2\text{O}$  Mixtures.



$\text{PH}_3 + \text{N}_2\text{O}$

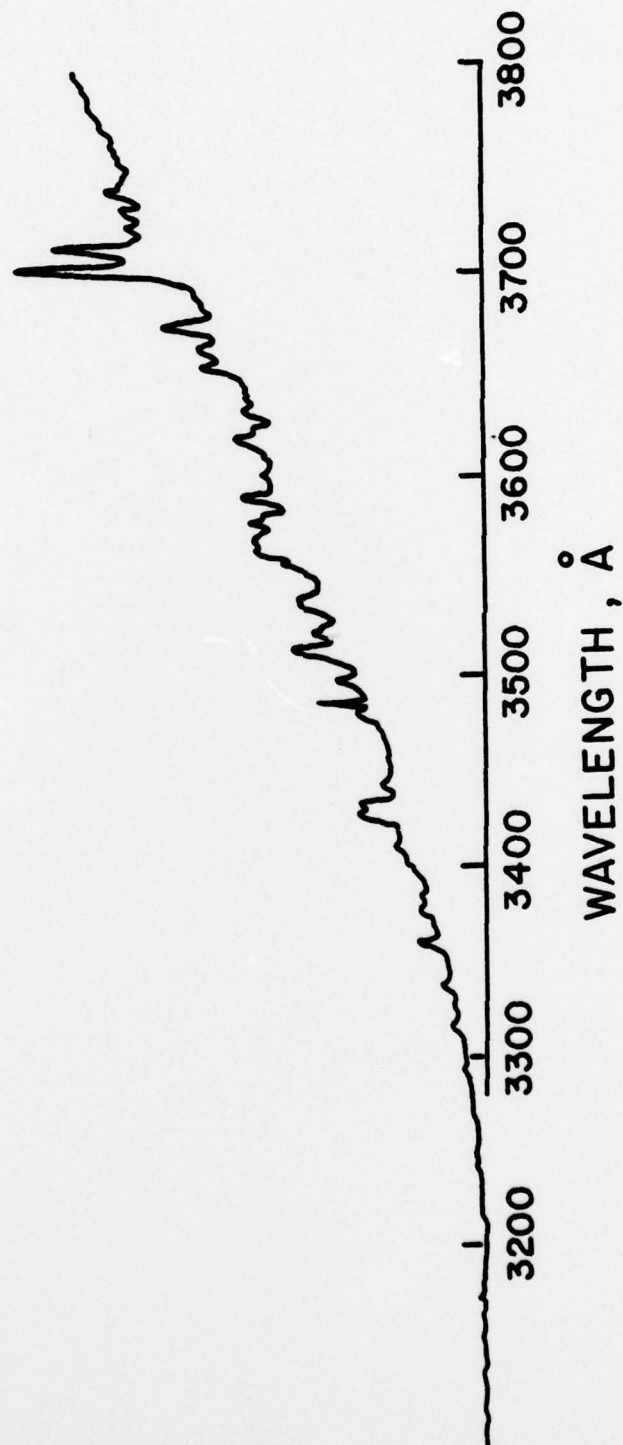


Figure 11: Chemiluminescence Spectra for  $\text{PH}_3/\text{N}_2\text{O}$  System.

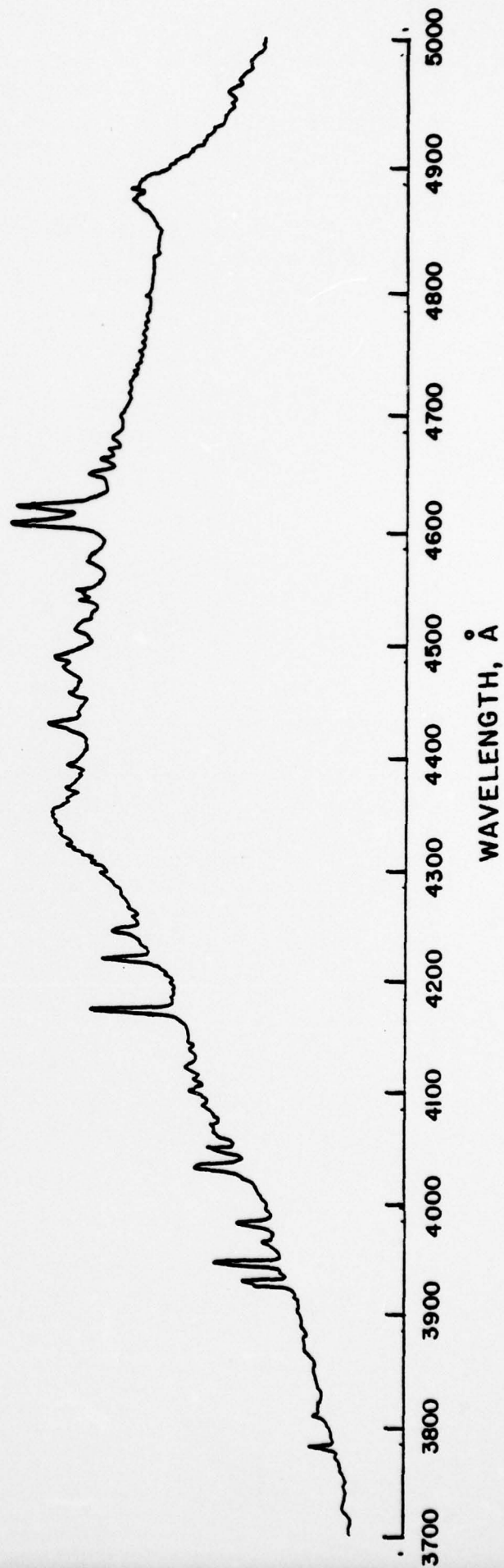


Figure 12: Chemiluminescence Spectra for  $\text{PH}_3/\text{N}_2\text{O}$  System.

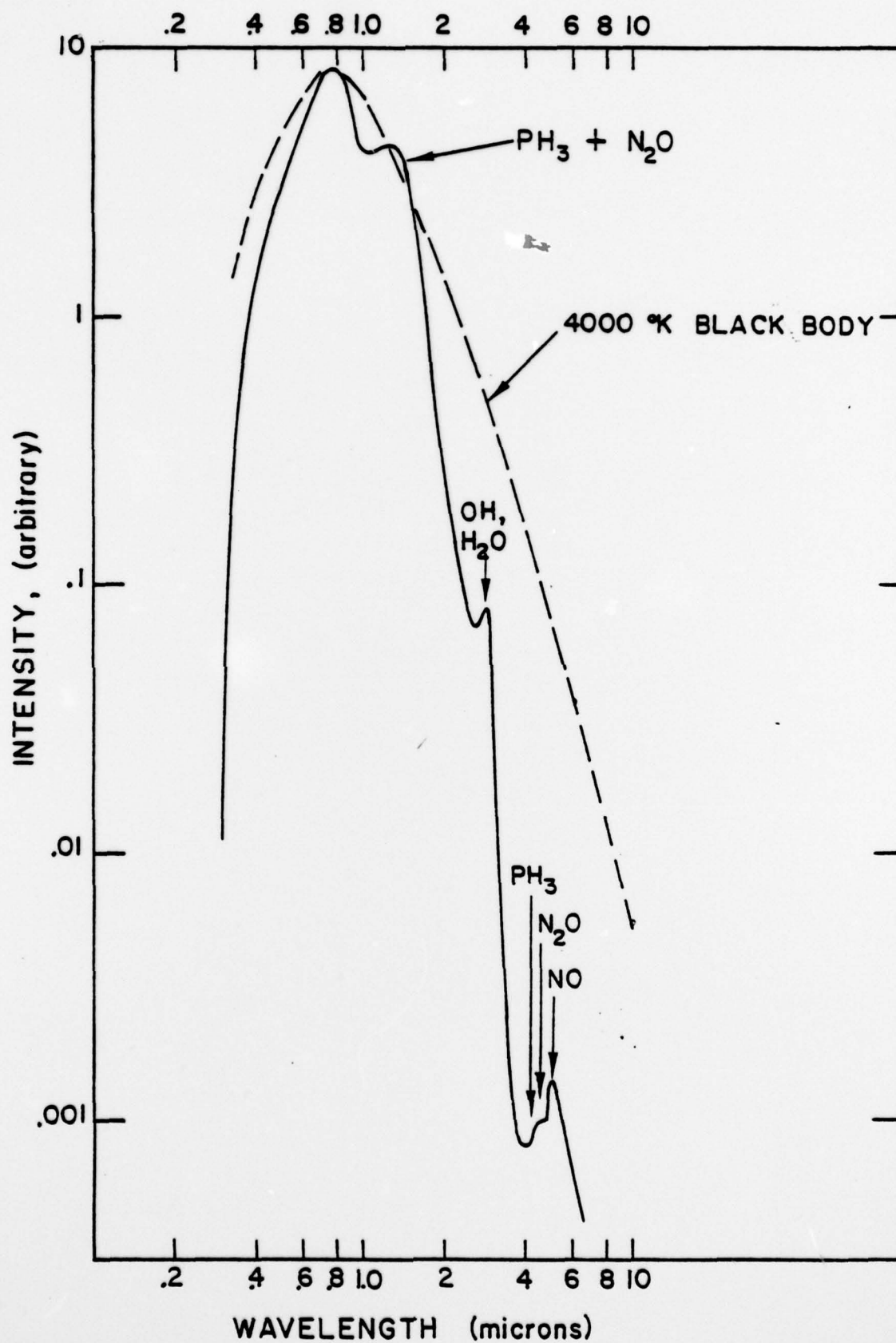


Figure 13: Absolute  
Emission Intensity Data  
for  $\text{PH}_3/\text{N}_2\text{O}$  System.



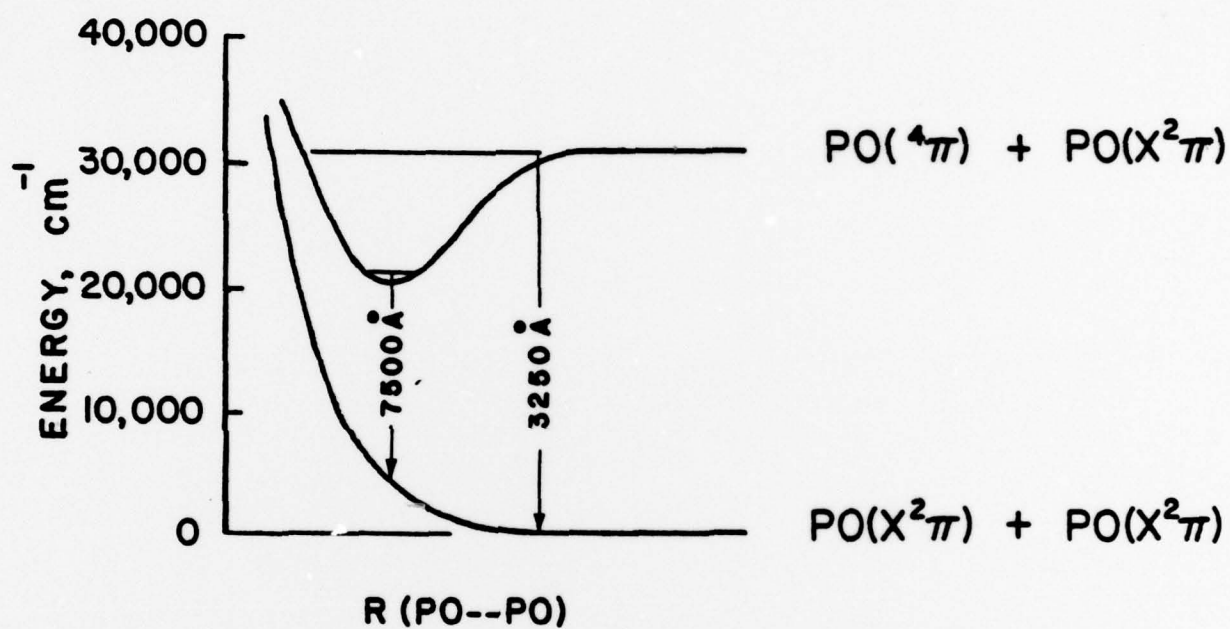
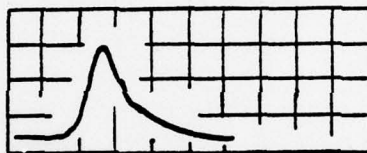
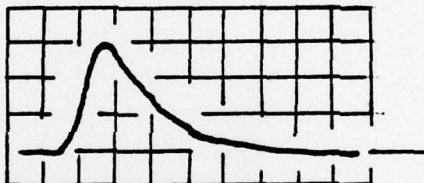


Figure 14: Schematic Cross Section of Potential Energy Hypersurfaces for  $(\text{PO})_2$  Dimer.

DYE LASER, 5900 Å



EXCIMER EMISSION



1  $\mu$ sec/div

Figure 15: Temporal Profiles of Dye Laser Pulse and  $\text{PH}_3/\text{N}_2\text{O}$  Spontaneous Emission.

## SPONTANEOUS EMISSION

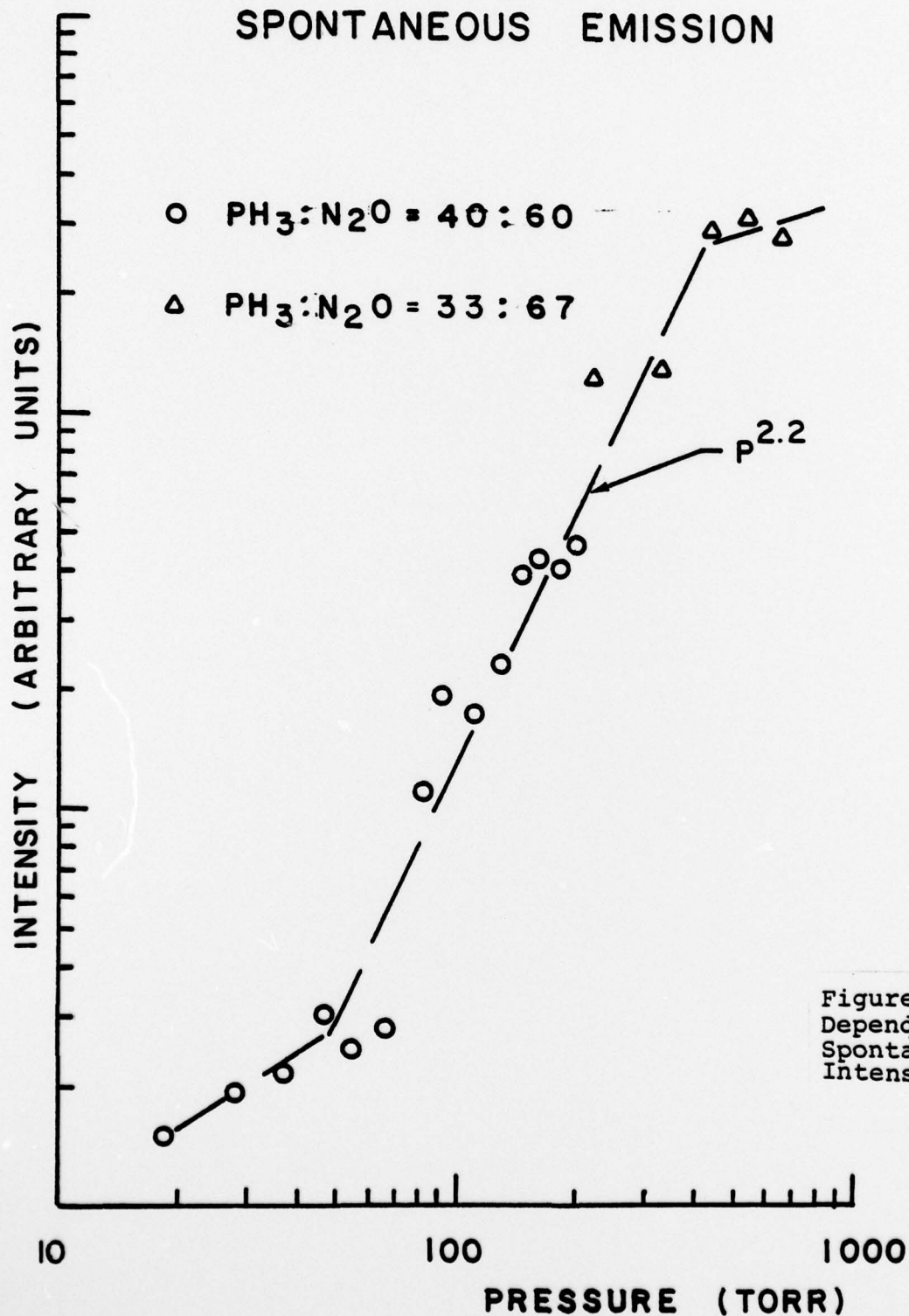


Figure 16: Pressure Dependence of  $\text{PH}_3/\text{N}_2\text{O}$  Spontaneous Emission Intensity.

lower trace of Figure 15 is plotted as a function of initial total reagent pressure. For pressures above about 20 Torr the temporal profile remains constant in duration, but the peak intensity varies approximately as the square of the pressure up to pressures of one atmosphere. Our cell is not designed for operation at higher pressures, but it is clear that quenching processes do not become dominant for pressures below one atmosphere.

Additional work on this interesting reaction system will be continued in an attempt to positively identify the emitter responsible for the strong radiation we observe.



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PUBLICATIONS

A cumulative list of publications for work supported under Grant AFOSR-73-2550 is given below.

1. K. K. Hui and T. A. Cool, "Experiments Concerning the Laser Enhanced Reaction Between Vibrationally Excited  $O_3$  and NO", to be published, J. Chem. Phys.
2. T. A. Cool, "Chemical Lasers", to be published as Chapter 5 in Dynamics and Energetics of Elementary Reactions, I. W. W. Smith, Editor, Plenum Publishing Company.
3. D. R. Fischell and T. A. Cool, "Spontaneous Emission from ScF in a Supersonic Mixing Flame", Proceedings of the Third Summer Colloquium on Electronic Transition Lasers, Snowmass, Colorado, September, 1976.
4. M. S. Chou and T. A. Cool, "New Metal Atom Transitions in As, Bi, Cu, Ge, Hg, In, Pb, Sb, and Tl", Proceedings of the Second Summer Colloquium on Electronic Transition Lasers, Woods Hole, Mass., September, 1975.
5. J. S. Whittier and T. A. Cool, "Initiation with an Electron Beam of Chemical Reactions of Interest for Visible Wavelength Lasers", Proceedings of the Second Summer Colloquium on Electronic Transition Lasers, Woods Hole, Mass., September, 1975.
6. J. A. McGarvey, N. E. Friedman, and T. A. Cool, "Vibrational Energy Transfer in Hf-HCN, DF-HCN, and  $H_2$ -HCN Mixtures from 240-450°K", J. Chem. Phys., 66, 3189 (1977).

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